Ferroelectrics

ISSN: 0015-0193 (Print) 1563-5112 (Online) Journal homepage: https://www.tandfonline.com/loi/gfer20

Physical properties and reentrant behavior in PLZT thin films

M. Melo, E.B. Araujo, E.A. Neradovskaya, A.P. Turygin, A.A. Esin, V.Ya. Shur & A.L. Kholkin

To cite this article: M. Melo, E.B. Araujo, E.A. Neradovskaya, A.P. Turygin, A.A. Esin, V.Ya. Shur & A.L. Kholkin (2017) Physical properties and reentrant behavior in PLZT thin films, Ferroelectrics, 509:1, 1-9, DOI: 10.1080/00150193.2017.1289792

To link to this article: https://doi.org/10.1080/00150193.2017.1289792

Published online: 10 May 2017.

FERROELECTRICS
2017, VOL. 509, 1–9
http://dx.doi.org/10.1080/00150193.2017.1289792

Taylor & Francis
Taylor & Francis Group

Physical properties and reentrant behavior in PLZT thin films

M. Melo[a], E.B. Araujo[a], E.A. Neradovskaya[b], A.P. Turygin[b], A.A. Esin[b], V.Ya. Shur[b], and A.L. Kholkin[b,c]

[a]Department of Physics and Chemistry, São Paulo State University, Ilha Solteira, SP, Brazil; [b]School of Natural Sciences and Mathematics, Ural Federal University, Ekaterinburg, Russia; [c]Department of Physics & CICECO – Aveiro Institute of Materials, University of Aveiro, Aveiro, Portugal

ABSTRACT

La-modified lead zirconate titanate (PLZT) thin films were prepared to study their physical properties at macro- and nanoscale. Piezoresponse force microscopy (PFM) studies suggest a local imprint behavior at room temperature. Confirmed by P-E hysteresis loops recorded at 180-300 K, the imprint effect at room temperature tends to disappear at lower temperatures. In addition, the remanent polarization gradually increases and then decreases after reaching a maximum at around 243 K. This behavior suggests the occurrence of a reentrant dipole glass or an activated electric field effect in the studied PLZT films.

ARTICLE HISTORY
Received 29 August 2016
Accepted 21 November 2016

KEYWORDS
PLZT; thin films; relaxors; dipole glass

1. Introduction

Lanthanum-modified lead zirconate titanate $Pb_{1-x}La_x(Zr_{1-y}Ti_y)_{1-x/4}O_3$ (PLZT) in the ceramic form has been used in different technological applications specially as a result of their remarkable electrooptic properties [1, 2]. Depending on the La/Zr/Ti composition, PLZT exhibits either ferroelectric or relaxor behavior. Relaxor behavior in PLZT is related to the charge disorder caused by cations of different valences randomly distributed over the equivalent crystallographic positions, i.e., Pb^{2+} and La^{3+} on A sites of the perovskite structure [3]. In contrast to normal ferroelectrics, a wide peak in the temperature dependence of the dielectric permittivity with temperature and frequency dispersion characterizes the relaxor phenomenon. In addition, the phase transition in these materials depends on the external applied electric field on cooling, so that the material transforms from a paraelectric phase to an ergodic relaxor one [3]. From the technological point of view, PLZT composition La/Zr/Ti = 9/65/35 mol% is particularly important since its dielectric permittivity and electro-optical coefficients exhibit their maximum values at this ratio [4, 5].

In the past two decades, there was an extensive research on PLZT thin films mainly motivated by the advances in instrumentation to probe physical properties with nanoscale resolution [6], by the advent of new integrated optical devices [7], and by the better scientific comprehension of their physical properties in comparison with bulk ceramics [8]. In addition to open issues associated to the nature of the polar nano-clusters in the physics of relaxors [9] or to the new strategies to map the local polarization disorder and associated

CONTACT E.B. Araujo eudes@dfq.feis.unesp.br

properties at nanoscale in PLZT system [10], other issues have received attention of the scientific community. Among these issues, a reentrant dipole glass feature recently observed in perovskite-type $BaTiO_3$-$BiScO_3$ [11, 12] and $BaTiO_3$-$AgNbO_3$ [13] solid solutions opened fruitful discussions on the possible scenarios giving rise to such phenomenon. A gradual increase in the switchable polarization on cooling up to a maximum value before decreasing the polarization at lower temperatures characterizes the reentrant behavior in these materials [12]. On the other hand, a recent report on the like reentrant relaxor behavior observed in tungsten bronze $(Sr,Ba)Nb_2O_6$ (SBN) ceramics suggests that the observed phenomenon is due to the increase of coercive field rather than a decrease in the long-range interaction of structural dipoles on cooling [14]. This scenario demonstrates that the subject is open for discussions and has motivated additional investigations on the origin of the reentrant phenomena in relaxors.

In the present work, we investigate the local piezoelectric properties and the effects of frequency and temperature on the coercive field and remanent polarization of polycrystalline PLZT thin films prepared by chemical route. To our knowledge, there are no reports in the literature addressing the issue in ceramics or in PLZT thin films. The local piezoelectric properties were studied using piezoresponse force microscopy (PFM) technique at room temperature, while the macroscopic P-E hysteresis loops were acquired to access the ferroelectric properties at different temperatures and frequencies.

2. Experimental

PLZT thin films with nominal composition $(Pb_{0.91}La_{0.09})(Zr_{0.65}Ti_{0.35})_{0.977}O_3$ have been deposited on $Pt/TiO_2/SiO_2/Si(100)$ substrates by spin coating at 5000 rpm for 30 seconds using a polymeric chemical route based on modified Pechini method, such as described elsewhere [15]. The pyrolysis was carried out by putting the films directly on a hot plate at ~200°C for 5 minutes and then in an electric furnace at 300 °C for 1 hour. The film thickness increases when depositing new layers on the previously pyrolyzed film layer and repeating the same procedure to remove the organics. Upon obtaining the desired film thickness, a final temperature annealing at 700 °C for 1 hour promoted the final film crystallization. The obtained films were 540 nm in thickness.

PFM using a commercial atomic force microscope (Ntegra Prima, NT-MDT) equipped with a tip-cantilever system (Nanosensors) has been used to study the local piezoelectric properties of the PLZT films. The PFM images were acquired under a probing AC voltage with amplitude of 5 V and frequency of 50 kHz. The driving frequency was chosen being far from contact resonance in the cantilever-sample system to avoid the ambiguity in experimental data. The PFM signal was acquired by a lock-in amplifier (SR-830A, Stanford Research) set for the time constant and sensitivity of 10 ms and 1 mV, respectively. The PFM images were obtained in vertical PFM mode ($R\cos\varphi$) so that bright and dark contrast reflected the parallel or antiparallel polarization directions caused by the probing electric field [16].

For electrical measurements, the top electrodes were sputter-deposited as an array of Au circular dots (0.3 mm in diameter) over an area of 1×1 cm^2. P-E hysteresis loops were measured using FE module of AixACCT TF 2000 system (aixACCT Systems GmbH, Germany). Bipolar triangular voltage pulses were applied to the sample and the current was measured using a virtual ground transimpedance amplifier. The prepolarization pulses were

established at a defined polarization state before each measurement. The sample was mounted on the Linkam THMS 600 (Linkam Scientific) stage to maintain the temperature in the range from RT to −90 °C. The probe station with silver tips (curvature radius 70 μm) was used to provide an electrical contact with the samples. The contact pressure was finely adjusted to maintain a good electrical contact without film puncture.

3. Results and discussion

The topography and piezoresponse images of the studied PLZT thin film are shown in Figs. 1a and 1b, respectively. Although some elongated grains were observed in topography image, we saw mainly spherical grains with the average grain size 438 nm. The bright and dark areas in out-of-plane image in Fig. 1b refer to upward and downward polarization states, respectively, while intermediate contrast refers to piezoresponse signal close to zero. The homogeneous contrast in Fig. 1b reveals that studied film is quite homogeneous in terms of piezoresponse.

Black squares in Fig. 1c refer to the distribution of the piezoresponse signal acquired from piezoresponse image in Fig. 1b, while red line in the same figure is a fitting curve performed using a Gaussian function. The curve similar to the one shown in Fig. 1c is a statistical distribution of the piezoelectric signal related to the domain configurations in a ferroelectric [17],

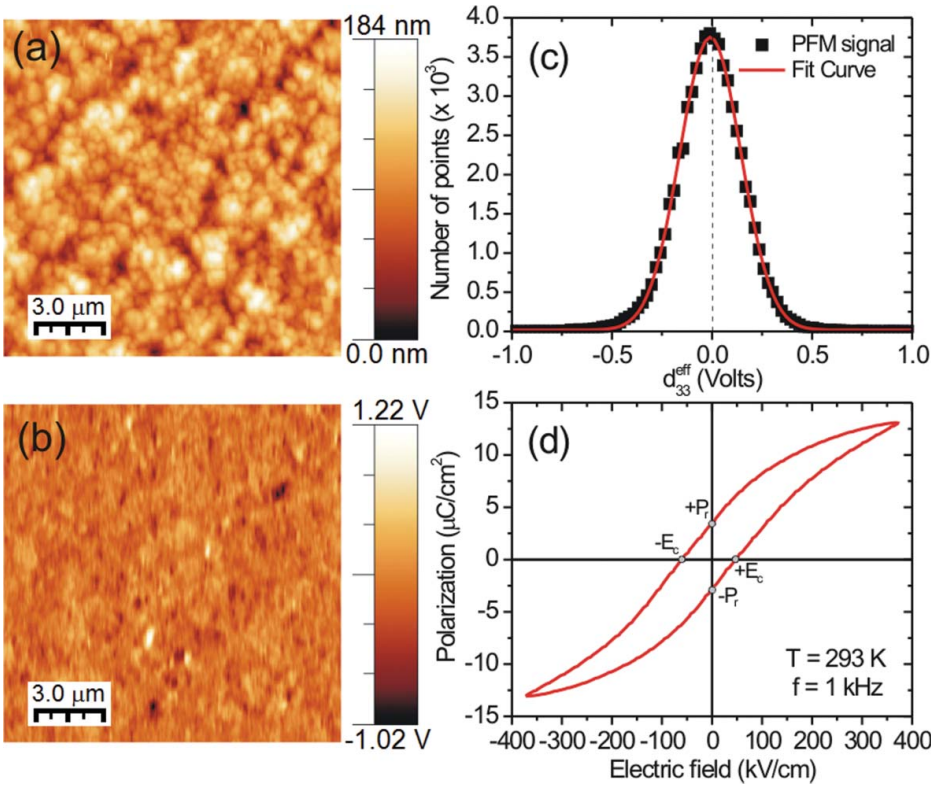

Figure 1. (a) Topography and (b) piezoresponse images (15 × 15 μm^2) of the studied PLZT thin film. (c) Distribution of the local piezoresponse signal and (d) hysteresis loops at 1 kHz frequency and 293 K.

where the out-of-plane polarization is due to the effective d_{33} piezoelectric coefficient [16]. Thus, the observed peak in this distribution curve is associated with the most probable domain configuration. As such, only one peak observed in Fig. 1c indicates predominance of unimodal polarization domains in the studied PLZT film.

In Fig. 1c, the d_{33} peak is slightly shifted to positive values (d_{33} peak $\sim 5.7 \times 10^{-3}$ V) and is almost symmetrical around 0 V. Let us now consider the relevance of the peak shift and asymmetries in the distribution curve shown in Fig. 1c. These peak shifts or asymmetries may be due to instrumental effects or be a result of self-polarization (an imprint effect) in the films. In the present study, we used stiff cantilevers with k spring constant interacting with film surface, so that the capacitive contribution ($\propto V_{ac}/k$) of the cantilever is expected to be minimal if compared to the piezoelectric contribution ($d_{33}V_{ac}$) [18]. Under these considerations and the additional fact that the magnitude of the effective piezoelectric coefficient d_{33} is independent on the tip radius [19], tip geometry or electrostatic effects can be excluded as probable mechanisms responsible by small asymmetry observed in the distribution curve in Fig. 1c.

Excluding instrumental effects, imprint is another possible effect responsible for the small shift observed on the d_{33} peak in distribution of the piezoresponse signal. Sometimes referred as a self-polarization effect [20], the imprint appears typically in ferroelectric thin films due to an internal built-in electric field leading to asymmetries in the hysteresis loops at both macro- and nanoscale. To infer about a possible self-polarization in the studied film, Fig. 1d plots macroscale P-E hysteresis loops at 1 kHz frequency at room temperature (293 K). The coercive field and remanent polarization are marked in the figure. The asymmetries observed in remanent polarization ($\Delta P_r = P_r^+ - P_r^- = +0.7\ \mu C/cm^2$) and coercive field ($\Delta E_c = E_c^+ - E_c^- = -12.8$ kV/cm) suggest a weak self-polarization with a predominance of domains toward the free surface of the film. In other words, $\Delta E_c < 0$ and $\Delta P_r > 0$ assume a small build-in electric field toward free surface of the film. Assuming a qualitative description for the self-polarization (P_i) described by the following $P_i \propto d_{33}$ [21], where d_{33} is the piezoresponse signal at maximum in the distribution curve in Fig. 1c, the asymmetry observed in P-E hysteresis loops in Fig. 1d agrees with the small shift observed in the effective d_{33} in Fig. 1c.

Fig. 2a shows the P-E hysteresis loops in PLZT thin film at 183 K in the frequency range from 50 Hz to 5 kHz, while the hysteresis loops at 1 kHz frequency at temperatures in the range 183–293 K are shown in Fig. 2b. Small changes were observed in the saturation polarization in both figures, as well as weak changes in the remanent polarization and coercive field. To reveal the temperature dependence of hysteresis loops, the differences in the remanent polarizations ($\Delta P_r = P_r^+ - P_r^-$) and coercive fields ($\Delta E_c = E_c^+ - E_c^-$) as functions of temperature were plotted in Figs. 2c and 2d, respectively.

From Figs. 2c and 2d, we observe that both ΔP_r and ΔE_c decrease with temperature decreasing indicating more symmetrical hysteresis loops at low temperatures. The thickness dependence of the self-polarization of PLZT films recently reported [22] demonstrates that self-polarization increases for thinner films. Additionally, the results indicate that Schottky barriers close to the bottom film-substrate interface is the probable mechanism responsible for the self-polarization in the studied PLZT films. Applying an electric field on cooling, a diffuse phase transition relaxor (ergodic) to relaxor (nonergodic) or relaxor-to-ferroelectric phase transition are different possibilities to occur [3]. In both cases, it is expected that a build-in electric field will produce the same effect in the hysteresis loops in relaxor or

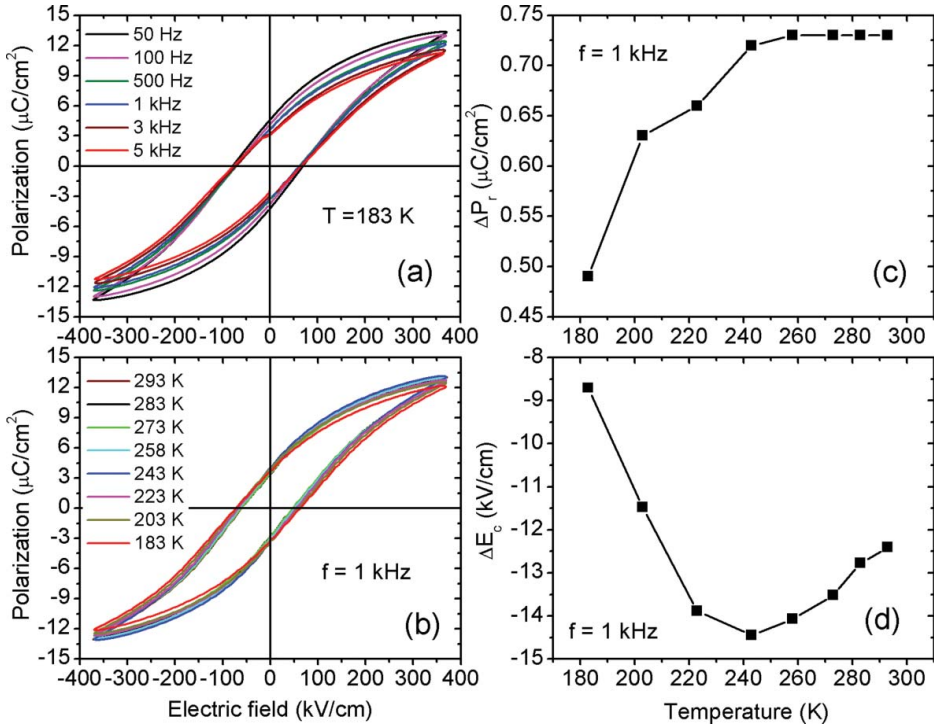

Figure 2. P-E hysteresis loops in PLZT thin film: (a) at 183 K in frequency ranged from 50 Hz to 5 kHz and (b) at 1 kHz frequency in temperature range from 183 to 293 K. Temperature dependences of the differences (c) $\Delta P_r = P_r^+ - P_r^-$ and (d) $\Delta E_c = E_c^+ - E_c^-$ at 1 kHz frequency. Lines in (c) and (d) are drawn as a guide to the eye.

ferroelectric phase. Therefore, the decrease in both ΔP_r and ΔE_c indicates that the build-in electric field in the bottom interface tends to vanish at low temperatures.

Figs. 3a and 3b show the frequency dependences of the remanent polarization and coercive field at the temperature 183 K, respectively. We observe that the remanent polarization decreases with increasing frequency within the studied range, while the coercive field decreases at first, and then increases almost linearly after reaching the minimum value at 500 Hz frequency. Often, the shape of the hysteresis loops can be affected by factors such as film thickness, oxygen vacancies, complex defects, mechanical stresses, preparation conditions, frequency of measurements, etc. [23]. Therefore, these factors also affect the related properties such as remanent polarization and coercive field.

The remanent polarization decreasing with the increase of frequency can be discussed in terms of domain motion. Increasing the frequency of the applied electric field, most domains are reversible by the external electric field, but some domains cannot follow the electric field during the switching. Consequently, the remanent polarization decreases with increasing frequency (Fig. 3a). In addition, defects and oxygen vacancies trapped in domain boundaries are responsible for the decrease of polarization in ferroelectric films, since pinned domains cannot be switched [24]. These mechanisms also lead to a decrease of the remanent polarization.

Based on the Avrami [25–27] theory for nucleation and crystallite growth, Ishibashi and Orihara [28] proposed a comprehensive model for the P-E hysteresis loops to explain the

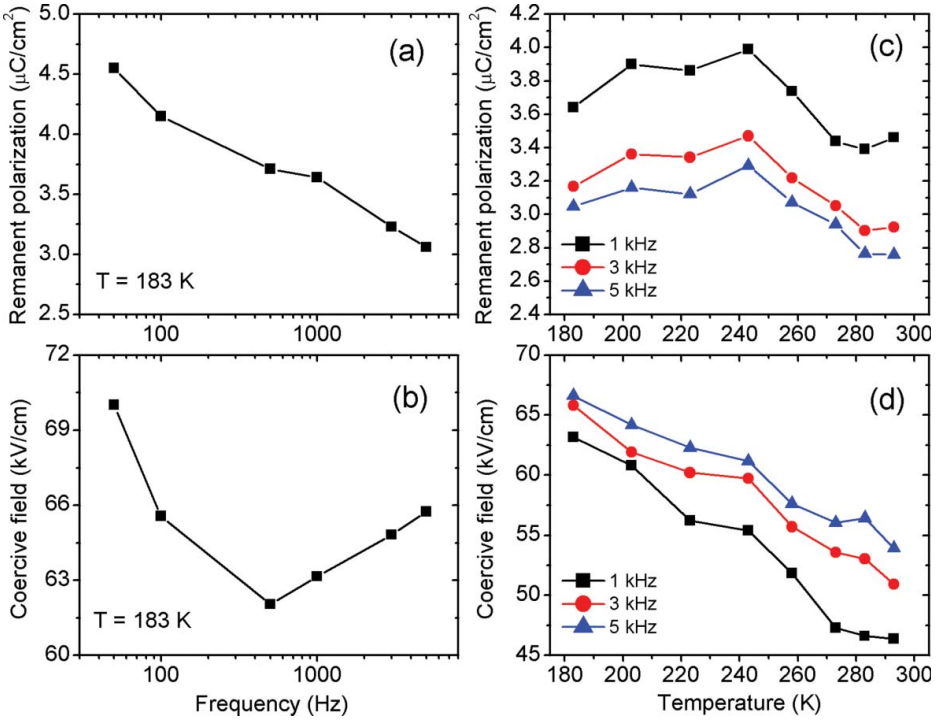

Figure 3. Frequency dependence of the (a) remanent polarization and (b) coercive field at 183 K. Temperature dependence of the (c) remanent polarization and (d) coercive field at 1, 3, and 5 kHz frequencies. Lines are drawn as a guide to the eye.

frequency dependence of coercive field in terms of a power law $E_c(f) \propto f^n$, where n is a frequency coefficient. Thus, the frequency behavior of coercive field is governed by the motion of domain walls in different ferroelectric systems and has been interpreted in terms of the resistance increasing to the domain wall motion during switching [29]. The above-mentioned power law for E_c has been experimentally observed for $Pb(Zr,Ti)O_3$ (PZT) ceramics [30] and thin films [31], and more recently has been theoretically studied in detail from the first principles [32]. However, the physics explaining the unexpected increase of coercive field observed in Fig. 3b with decreasing below 500 Hz is not yet clear. Higher E_c values observed at lower frequencies for Fe-doped PZT ceramics were attributed to the conduction contribution [33]. Despite the probable influence of defects and motion of the domain walls on the coercive field increasing at lower frequencies as observed in this work, further studies are needed to understand better this behavior.

Figs. 3c and 3d show the remanent polarization and coercive field behaviors as functions of temperature at three different frequencies. As the temperature decreases, the remanent polarization initially increases and then decreases slightly after reaching a maximum at 243 K for all frequencies. On the other hand, the coercive field increases almost linearly for all frequencies when temperature decreases. This trend suggests the occurrence of a reentrant dipole glass or an activated electric field effect in the studied PLZT film.

The dielectric relaxation in relaxors characterized by frequency dispersion in the temperature dependence of the dielectric permittivity is also observed in strongly disordered dipole glass materials [3]. By analogy with the spin-glass theory, the basic features of relaxors could

be explained [34], so that the transition from weak-ferroelectric to the non-ergodic dipole-glass state is a possible explanation for the reentrant dipole-glass behavior observed in the $BaTiO_3$-$BiScO_3$ [11] and $BaTiO_3$-$AgNbO_3$ [13] systems. More consistent with dipole glass behavior (as compared to canonical relaxor ferroelectric such as $Pb(Mg_{1/3},Nb_{2/3})O_3$ or uniaxial relaxor $(Sr,Ba)Nb_2O_6$), the characteristics, such as high activation energies (~0.2-0.3 eV) determined by Vogel-Fulcher relation and the maximum of P_r before it decreases at low temperatures, suggest a "true" reentrant phenomenon in the $BaTiO_3$-$BiScO_3$ system [11]. Other relaxor ferroelectric systems show similar behavior for the temperature dependence of the switchable polarization, but the origin of the phenomenon is attributed to other artifacts [14]. In a recent work, Bokov and Ye [35] have presented a comprehensive overview on the reentrant relaxors and concluded that in most cases it is difficult to distinguish between a genuine reentrant phenomenon and artifacts associated to intrinsic or extrinsic relaxation process.

PLZT system has been intensively studied in the past by scientific and technological reasons, but some topics remain open to discussions. Among rare reports on the field dependent switchable polarization on cooling, the effect of temperature on remanent polarization for PLZT compositions exhibits a gradual increase in P_r with decreasing temperature and no abrupt change at the dielectric permittivity maximum [36]. At the composition 9/65/35, this behavior has been associated with the fact that a metastable ferroelectric phase can be induced by application of sufficiently high electric field during cooling [3]. In addition, the temperature dependence of dielectric permittivity on field-cooled and zero-field-cooled-field-heated quasistatic regime indicates a glass-like freezing process in 9/65/35 PLZT ceramics [37]. Despite the temperature dependence of remanent polarization observed in the present work and the above considerations, it is not possible to affirm that it is a "true" reentrant dipole glass effect. Further investigations are, therefore, needed to examine the temperature dependence of remanent polarization under different electric fields to infer about the mechanisms and to clarify the true origin of this phenomenon observed in the present work.

4. Conclusion

In summary, physical properties of PLZT thin films at macro- and nanoscale were investigated. Studies using piezoresponse images indicate an existence of self-polarization at room temperature due to a build-in electric field so that domains are oriented towards film free surface. P-E hysteresis loops at different temperatures confirm the effect at macroscale that tends to disappear al low temperatures. The remanent polarization increasing on cooling and decreasing after reaching a maximum value at around 243 K suggests the occurrence of a reentrant dipole glass or an activated electric field effect in the studied PLZT film. However, it is not possible at this moment to affirm if this a "true" reentrant dipole glass effect or artifacts associated with intrinsic or extrinsic relaxation processes in the studied PLZT film. Further studies are needed to clarify the true origin of this phenomenon observed in the present work.

Funding

We would like to express our gratitude to the Brazilian agencies CNPq (Grant 304604/2015-1 and Project 400677/2014-8), CAPES and FAPESP for financial support. M.M. extends thanks to CNPq for

sandwich PhD fellowship (232241/2014-7). The work was partially supported by Russian Foundation for Basic Research (grant 16-02-00821-*a*) and Government of the Russian Federation (act 211, agreement 02.A03.21.0006), and also developed in the scope of Project CICECO-Aveiro Institute of Materials (ref. FCT UID/CTM/50011/2013), financed by national funds through the FCT/MEC and, when applicable, co-financed by FEDER under the PT2020 Partnership Agreement.

References

1. Haertling G. H., and Land C. E., Hot-pressed (Pb, La)(Zr,Ti)O_3 ferroelectric ceramics for electro-optic applications. *J Am Ceram Soc.* **54**, 1–11 (1971).
2. Haertling G. H., Electro-optic ceramics and devices. In: Levinson L. M., ed. *Electronic Ceramics*. New York: Marcel Dekker, 371–492 (1988).
3. Bokov A. A., and Ye Z. G., Recent progress in relaxor ferroelectrics with perovskite structure. *J Mat Sci.* **41**, 31–52 (2006).
4. Haertling G. H., Ferroelectric ceramics: History and technology. *J Am Ceram Soc.* **82**, 797–818 (1999).
5. Lynch C. S., The effect of uniaxial stress on the electro-mechanical response of 8/65/35 PLZT. *Acta Mater.* **44**, 4137–4148 (1996).
6. Ferri A., Saitzek S., Da Costa A., Desfeux R., Leclerc G., Bouregba R., and Poullain G., Thickness dependence of the nanoscale piezoelectric properties measured by piezoresponse force microscopy on (111)-oriented PLZT 10/40/60 thin films. *Surf Sci.* **602**, 1987–1992 (2008).
7. Fujii T., Suzuki T., Fujimori Y., Nakamura T., Moriwake M., and Takasu H., Development of (Pb, La)(Zr,Ti)O_3 electro-optic thin film for high-speed spatial light modulator. *Jpn J Appl Phys.* **45**, 7520–7524 (2006).
8. Dimos D., Warren W. L., Sinclair M. B., Tuttle B. A., and Schwartz R. W., Photoinduced hysteresis changes and optical storage in (Pb,La)(Zr,Ti)O_3 thin-films and ceramics. *J Appl Phys.* **76**, 4305–4315 (1994).
9. Blinc R., Laguta V. V., and Zalar B., Polar nanoclusters in relaxors. *J Mat Sci.* **41**, 27–30 (2006).
10. Kholkin A. L., Kiselev D. A., Bdikin I. K., Sternberg A., Dkhil B., Jesse S., Ovchinnikov O., and Kalinin S. V., Mapping disorder in polycrystalline relaxors: A piezoresponse force microscopy approach. *Materials.* **3**, 4860–4870 (2010).
11. Bharadwaja SSN, Kim J. R., Ogihara H., Cross L. E., Trolier-McKinstry S., and Randall C. A., Critical slowing down mechanism and reentrant dipole glass phenomena in (1-x)$BaTiO_3$-x$BiScO_3$ (0.1 <= x <= 0.4): The high energy density dielectrics. *Phys Rev B.* **83**, 024106 (2011).
12. Krayzman V., Levin I., Woicik J. C., and Bridges F., Correlated rattling-ion origins of dielectric properties in reentrant dipole glasses $BaTiO_3$-$BiScO_3$. *Appl Phys Lett.* **107**, 192903 (2015).
13. Lei C., and Ye Z. G., Re-entrant-like relaxor behaviour in the new 0.99$BaTiO_3$-0.01$AgNbO_3$ solid solution. *J Phys Condens Matter.* **20**, 232201 (2008).
14. Huang C. J., Li K., Wu S. Y., Zhu X. L., and Chen X. M., Variation of ferroelectric hysteresis loop with temperature in (Sr_xBa_{1-x})Nb_2O_6 unfilled tungsten bronze ceramics. *J Materiomics.* **1**, 146–152 (2015).
15. Araujo E. B., Nahime B. O., Melo M., Dinelli F., Tantussi F., Baschieri P., Fuso F., and Allegrini M., Processing and structural properties of random oriented lead lanthanum zirconate titanate thin films. *Mater Res Bull.* **61**, 26–31 (2015).
16. Balke N., Bdikin I., Kalinin S. V., and Kholkin A. L., Electromechanical imaging and spectroscopy of ferroelectric and piezoelectric materials: State of the art and prospects for the future. *J Am Ceram Soc.* **92**, 1629–1647 (2009).
17. Wu A., Vilarinho P. M., Shvartsman V. V., Suchaneck G., and Kholkin A. L., Domain populations in lead zirconate titanate thin films of different compositions via piezoresponse force microscopy. *Nanotechnology.* **16**, 2587–2595 (2005).
18. Hong S., Woo J., Shin H., Jeon J. U., Pak Y. E., Colla E. L., Setter N., Kim E., and No K., Principle of ferroelectric domain imaging using atomic force microscope. *J Appl Phys.* **89**, 1377–1386 (2001).

19. Tian L., Vasudevarao A., Morozovska A. N., Eliseev E. A., Kalinin S. V., and Gopalan V., Nanoscale polarization profile across a 180 degrees ferroelectric domain wall extracted by quantitative piezoelectric force microscopy. *J Appl Phys.* **104**, 074110 (2008).
20. Afanasjev V. P., Petrov A. A., Pronin I. P., Tarakanov E. A., Kaptelov E. J., and Graul J., Polarization and self-polarization in thin $PbZr_{1-x}Ti_xO_3$ (PZT) films. *J Phys Condens Matter.* **13**, 8755–8763 (2001).
21. Lima E. C., Araujo E. B., Souza Filho A. G., Paschoal A. R., Bdikin I. K., and Kholkin A. L., Structural depth profile and nanoscale piezoelectric properties of randomly oriented $Pb(Zr_{0.50}Ti_{0.50})O_3$ thin films. *J Phys D Appl Phys.* **45**, 215304 (2012).
22. Melo M., Araújo E. B., Shvartsman V. V., Shur V. Y., and Kholkin A. L., Thickness effect on the structure, grain size, and local piezoresponse of self-polarized lead lanthanum zirconate titanate thin films. *J Appl Phys.* **120**, 054101 (2016).
23. Damjanovic D., Hysteresis in piezoelectric and ferroelectric materials. In: Bertotti G., Mayergoyz I. D., eds. *The science of hysteresis*. Oxford: Elsevier; 337–465 (2005).
24. Guo D., Wang C., Shen Q., Zhang L., Li M., and Liu J., Effect of measuring factors on ferroelectric properties of $Bi_{3.15}Nd_{0.85}Ti_3O_{12}$ thin films prepared by sol-gel method for non-volatile memory. *Appl Phys A*. **97**, 877–881 (2009).
25. Avrami M., Kinetics of phase change I - General theory. *J Chem Phys.* **7**, 1103–1112 (1939).
26. Avrami M., Kinetics of Phase Change. II Transformation-time relations for random distribution of nuclei. *J Chem Phys.* **8**, 212–224 (1940).
27. Avrami M., Granulation, phase change, and microstructure - Kinetics of phase change III. *J Chem Phys.* **9**, 177–184 (1941).
28. Ishibashi Y., and Orihara H., A theory of d-e hysteresis loop - application of Avrami model. *Integr Ferroelectr.* **9**, 57–61 (1995).
29. Scott J. F., *Ferroelectric memories*. Heidelberg: Springer (2000).
30. Lente M. H., Picinin A., Rino J. P., and Eiras J. A., 90 degrees domain wall relaxation and frequency dependence of the coercive field in the ferroelectric switching process. *J Appl Phys.* **95**, 2646–2653 (2004).
31. Yang S. M., Jo Y. J., Kim T. H., Yoon J. G., Song T. K., Lee H. N., Marton Z., Park S., Jo Y., and Noh T. W., Ac dynamics of ferroelectric domains from an investigation of the frequency dependence of hysteresis loops. *Phys Rev B.* **82**, 174125 (2010).
32. Liu S., Grinberg I., and Rappe A. M., Intrinsic ferroelectric switching from first principles. *Nature.* **534**, 360–363 (2016).
33. Lente M. H., and Eiras J. A., Frequency dependence of the switching polarisation in PZT ceramics. *Ferroelectrics.* **257**, 227–232 (2001).
34. Timonin P. N., Dipole-glass concept and history-dependent phenomena in relaxors. *Ferroelectrics.* **400**, 427–433 (2010).
35. Bokov A. A., and Ye Z. G., Reentrant phenomena in relaxors. In: Algueró M., Gregg J. M., Mitoseriu L., eds. *Nanoscale ferroelectrics and multiferroics: Key processing and characterization issues, and nanoscale effects.* Chichester: John Wiley & Sons; 729–764 (2016).
36. G. H. Haertling, Improved hot-pressed electrooptic ceramics in the $(Pb,La)(Zr,Ti)O_3$ system. *J Am Ceram Soc.* **54**, 303–309 (1971).
37. Kutnjak Z., Bobnar V., Filipic C., and Levstik A., Glassy properties of 9/65/35 PLZT ceramics. *Ferroelectrics.* **257**, 29–38 (2001).